# THE OPTIMUM PARTICLE-SIZE DISTRIBUTION OF COAL FOR COAL-WATER SLURRIES

C.B. Henderson and R.S. Scheffee

Atlantic Research Corporation 5390 Cherokee Avenue Alexandria, Virginia 22314

#### ABSTRACT

The development of coal-water slurries as a boiler fuel is rapidly approaching commercialization. As distinguished from dilute pipeline slurries, fuel slurries are stable, highly-loaded (e.g. 70% dry coal by weight) liquids which can be transported, stored, pumped, and burned like residual fuel oil. One of the key technologies required to obtain highly-loaded, low-viscosity mixtures is the particle size distribution of the coal. Two derivations of the optimum particle-size distribution for minimum slurry viscosity are compared and shown to result in equivalent forms. The theory of grinding is reviewed, and calculated distributions are presented. Experimentally-determined particle-size distributions obtained by both wet and dry grinding in ball mills are given and compared with the theoretical grinding results. It is shown that near-optimum distributions can be obtained by blending.

#### INTRODUCTION

For several years, the Atlantic Research Corporation has been developing coal-water slurries as a low-cost liquid boiler fuel. As distinguished from dilute pipeline slurries, fuel slurries are stable, highly-loaded (e.g. 70% dry coal by weight) liquids which can be transported, stored, pumped, and burned like residual fuel oil. To achieve the required rheological properties in highly-loaded suspensions, requires a very carefully selected distribution of particle sizes and the incorporation of surface-active additives. The subject of particle-size distribution, its optimization and control, is the subject of this paper.

#### II. OPTIMUM PARTICLE-SIZE DISTRIBUTION

The rheology of a suspension of particles within a liquid is influenced by a number of factors which include the size and the distribution of sizes of the particles, particle concentration, particle shape, and surface charge. The effects of surface properties, although often very important, do not fall within the scope of this paper.

The sizing of particles to obtain minimum viscosity of a suspension is closely related to the sizing of particles for maximum packing density of powder beds. If we ignore liquid-particle and particle-particle surface effects, the particle-size distribution resulting in maximum bed density will also result in minimum viscosity for a specified particle concentration. Consequently, the large body of literature addressing both suspensions and powder beds can be utilized. A review of the extensive literature will not be addressed here; only the work of three authors will be discussed. All of the work will be limited to either ideal powder beds consisting of spherical particles with only mechanical interactions or ideal suspensions containing spherical particles and within which all surface interactions are absent.

In 1928, Furnas (1) derived a theory for the maximum packing density of bimodal mixtures, i.e. a bed containing particles of two discrete particle diameters. Assuming that the fractional void volumes,  $\boldsymbol{\nu}$ , of the individual monomodal beds are identical and that the ratio of the two particle diameters is large, the composition of maximum density

is obtained if the small particles fill the interstices of the large particles such that the total bed volume does not increase. In a unit cell, the absolute volume of large particles is  $V_{0}=1-\nu$ . Filling the void volume  $\nu$  with small particles, the absolute volume of small particles is  $V_{0}=\nu$  (1- $\nu$ ). The volume ratio of large particles to small particles is  $1/\nu$  and the void volume of the mixed bed is  $\nu^{2}$ . When the ratio of the two particle diameters is greater than about 50, the theory is in good agreement with experimental data. For ratios, less than 50, Furnas employed experimental data to obtain generalized relationships between voids in the bimodal bed, concentration of large particles, void fraction in the monomodal beds, and the particle diameter ratio. These relationships have been greatly utilized in many industries for discrete particle mixtures. Both theory and experiment have subsequently been extended to multimodal mixtures of discrete particles.

Andreasen and Andersen in 1930 (2) and Furnas in 1931 (3) extended the theory to continuously varying particle diameters. Andreasen and Andersen start by approximating the continuous distribution with a series of particle size fractions in which the particle diameters form a geometrical progression. Starting with the fine fraction they successively add the coarser fractions in such a way that the amount of each fraction added is proportional to the amount of undersize already present:

$$\delta v_{j} \sim \sum_{i=1}^{j-1} v_{i}$$
 (1)

where i = 1 designates the smallest size fraction

Reducing the constant ratio between size fractions to a differential amount, results in a differential equation:

$$\frac{d\ln V}{d\ln D} = n = constant, \tag{2}$$

which upon solution leads to a continuous particle size distribution:

$$\mathbf{F}_{\mathbf{p}} = \mathbf{c} \mathbf{D}^{\mathbf{n}} \tag{3}$$

where F = cumulative volume of particles smaller than D  $_{\rm c}^{\rm p}$  = constant

The somewhat arbitrary method by which this result is obtained is justified by comparison with experimental data which indicate that the exponent n should be in the range of 1/3 - 1/2.

Apparently independently, Furnas in 1931 arrived at a similar but somewhat more general result. He also approximates the continuous distribution as a geometrical series of discrete particle diameters. Designating the diameter of the largest particles as Df, the smallest as Dg, and the size ratio of successive size fractions as q (Furnas used  $\sqrt{2}$ , but his derivation is applicable to any constant value), the size of each individual fraction is (starting from the finest fraction):

$$D_1 = D_s, D_2 = qD_s, \dots D_i = q^{i-1}D_s, \dots D_l = q^{N-1}D_s$$
 (4)

where N = number of component particle sizes

If q is very large (e.g. 50), the extension of bimodal theory to multimodal mixtures requires that the volume ratio of each size fraction to the next smallest size fraction, R, be a constant. Furnas makes the assumption\* that this theorem also holds for any q, however small, with the provision that the volume ratio of consecutive size fractions is

<sup>\*</sup> This can be shown to be equivalent to the basic assumption employed by Andreasen and Andersen

no longer the same as for the optimum mixture at large values of q. Based on this assumption, Furnas obtains his equation for the optimum particle size distribution which he writes as follows:

$$F_{p} = \frac{R^{\log D}_{i} - R^{\log D}_{s}}{R^{\log D}_{\ell} - R^{\log D}_{s}}$$
(5)

Furnas did not specify the base of his logarithm. By following his derivation, it is readily shown that the base is equal to q. Rewriting the preceeding equation and defining the log's as the natural logarithims:

$$F_{p} = \frac{R^{\ln D_{i}/\ln q} - R^{\ln D_{s}/\ln q}}{R^{\ln D_{l}/\ln q} - R^{\ln D_{s}/\ln q}}$$
(6)

Since  $a^{c \ln b} = b^{c \ln a}$  for any values of a, b, and c, the preceeding equation can also be written

$$\vec{F}_{p}) = \frac{\vec{D}_{i}^{n} - \vec{D}_{s}^{n}}{\vec{D}_{p}^{n} - \vec{D}_{s}^{n}}$$
 (7)

$$n = \ln r / \ln q \tag{8}$$

Furnas' Equation 7 is a more general form of the Andreasen equation, the latter being equivalent to the former for  $D_S=0$ . The similarity in form between the two equations is to be expected considering the equivalence in the basic assumptions.

R is equal to 1/p for particle fractions separated by large values of q, but as q becomes smaller, R must decrease. Furnas presents a method for obtaining this relationship. First, he considers the optimum discrete particle- size distribution in a particle-size range specified by upper and lower limits,  $D_{\rm S}$ , and  $D_{\rm S}$ , respectively. He maintains that there is an optimum number of size fractions within the specified range for maximum loading since the volume decrease on mixing particles of different sizes becomes less efficient as the particle diameters become closer together. He next assumes that the experimentally determined relationship between the volume decrease and the size ratio in a binary system is the same as the relationship between the overall volume decrease and the ratio between consecutive sizes in an N-component system. The optimum number of size fractions is obtained by minimizing the overall volume decrease on mixing the size fractions in the multicomponent system. Furnas is now ready to obtain his expression for R. He assumes that the volumetric ratio of the largest particles to the smallest particles should be the same for any N and therefore  $R^{N-1}$  sould be a constant independent of q, or equivalently of N. He obtains this constant from his optimum number of size fractions obtained for a specified size range. Combining results, the exponent n in Equation 8, is found to be a complex function of  $\nu$  and the ratio D<sub>f</sub> /D<sub>s</sub>. The first, and most important conclusion, is that n is independent of q. This means that the derivation is as valid for continuous distributions, as claimed by Furnas, as for discrete distributions. The validity is of course dependent upon the assumptions employed. For = y 0.4 and  $D_0/D_s$  = 500, the optimum value of n according to the Furnas analysis is calculated

Farris (4) presents his analysis in terms of the viscosity behavior of multimodal suspensions in a liquid instead of the maximum packing of a particle bed. He optimizes the particle size distribution to obtain the minimum viscosity for a given volumetric concentration of particles. He also makes an assumption which is equivalent to that made by Andreasen and Andersen as represented in Equation 1 and to that made by Furnas in deriving Equation 5. Farris, however, presents his arguments more logically and with an improved basis of experimental data.

Farris noted that there were several references in the experimental literature to the fact that the finer particles in a bimodal suspension behave essentially as a fluid toward the coarser particles. To be more specific, if the ratio of particle sizes is large (e.g.

10 or greater), the resistance to motion encountered by a large sphere is the same when passing through a suspension of smaller spheres as when it passes through a pure liquid of the same viscosity and density as the suspension.

Farris first starts with the relative viscosity (ratio of suspension viscosity to liquid viscosity) of a monomodal system, which for non-interacting spherical particles has been shown experimentally to be a unique function of the volume fraction of solids. He next adds coarser particles of much larger diameter (i.e. q is large) to the suspension. Based on the assumption that the fines act like the liquid, he calculates the relative viscosity of the bimodal mixture using the same form as for the monomodal system. By successively adding coarser fractions, each larger by a large factor than the next smaller fraction, he extends his analysis to multicomponent distributions.

Farris obtains the optimum particle size distribution by differentiating the viscosity of the suspension with respect to composition, equating to zero, and solving for the compositional variables. The optimum composition of a multicomponent system is expressed as follows:

$$\phi_{1} = \phi_{2} = \phi_{3} \dots = \phi_{N}$$
where: 
$$\phi_{i} = v_{i} / \frac{1}{\Sigma} v_{k}$$

$$v_{i} = \text{volume of } i^{\text{th}} \text{ component}$$

$$v_{o} = \text{volume of liquid}$$
(9)

The form of this series of N-l equations is perfectly satisfactory to define the optimum composition. As it stands, however, it is difficult to compare with other equations. If, however, the N-l equations are used to solve for N-l values of  $V_i$  in terms of an N<sup>th</sup> variable, a particle-size distribution can be constructed. It is convenient to select the particle-loading, g, defined as the volume of particles dividied by the total volume, as the N<sup>th</sup> variable. After considerable algebra, the final equation is as follows:

$$F_{p} = \frac{\Theta^{1-1} - 1}{\Theta^{N} - 1}$$
 (11)

where 
$$\theta = (1-g)^{-1/N}$$
 (12)

This distribution is independent of the size of the individual fractions. If we specify size fractions with a geometrical progression of particle diameters and minimum and maximum particle diameters  $D_{\rm S}$  and  $D_{\rm L}$ , respectively, the preceding equation is transformed to:

$$F_{p} = \frac{D^{n} - D_{s}^{n}}{D_{l}^{n} - D_{s}^{n}}$$
 (13)

where 
$$n = \frac{\ln (\frac{1}{1-g})}{\ln \frac{1}{1} \ln \frac{1}{g}}$$
 (14)

We note that the form of the distribution equation, Eq (14), is exactly the same as that given by Furnas, Eq. (7). The exponent differs, however. For g = 0.65 and  $D_{\mu}/D_{s}$  = 500, a value of 0.17 is calculated for n.

The derivation just described is based on particle-size fractions separated by large factors, that is by large values of q. Farris claims that his results, i.e. Eqns. (9) and (10) are also applicable in systems in which there is an equal interaction between

particles of a different size. For interacting particles, Farris defines a crowding factor f as the fraction of one size that behaves as if it were the next largest size. For spherical particles that interact only mechanically: similarity should apply, f should be equal for all particle sizes, and Equation 9 will be applicable. Applying a definition of f and proceeding through considerable algebra, it can be shown that Equations 13 and 14 are still pertinent and independent of the value of f.

Equations 13 and 14 were derived starting with a discontinuous particle-size distribution. In arriving at the final form, the factor q, denoting the separation in size between successive fractions, does not appear. Consequently, it can be concluded that the results are equally applicable to continuous distributions.

## III. EFFECT OF PARTICLE-SIZE DISTRIBUTION ON VISCOSITY

In the preceeding section, the concept that fine particles in a suspension behave as a fluid toward coarser particles was used to obtain an optimum particle- size distribution. This same concept was also used by Farris to calculate the viscosity of multimodal suspensions provided the particle size fractions are separated by large factors. These calculations can be extended to suspensions of mechanically interacting particles (i.e., the individual size fractions are separated by small factors), provided the crowding factors are known - presumably obtained experimentally.

The viscosity of coal-water suspensions is greatly influenced by surface chemistry, which is outside the scope of this paper. Consequently, the viscosities calculated ignoring these effects are of limited value, and the principal value of these calculations is to illustrate qualitatively the effect of particle- size distribution on viscosity. The viscosity of a monomodal system is shown as a function of volume fraction of solids in Figure 1. This relationship has been obtained experimentally for noninteracting rigid spheres and is valid for particles as small as  $4\mu$  m and as large as 250  $\mu$  m. As the volume fraction of solids approaches 0.605, the relative viscosity (the ratio of the viscosity of the suspension, to that of the carrier) tends to very high numbers. This volume fraction corresponds approximately to a slurry consisting of 65% by weight of coal in water. Also shown are the relative viscosities of an optimal bimodal suspension. The reduction in viscosity by using a bimodal suspension is apparent. Additional reductions are obtained by increasing modality even further; for a volume fraction of 0.66 (corresponding to a 70% slurry of coal in water), relative viscosities of infinity, 51, 30, and 23 are calculated for monomodal, bimodal, trimodal, and tetramodal mixtures, respectively.

## IV. PARTICLE-SIZE DISTRIBUTIONS OBTAINED IN BALL MILLS

Coal as ordered from the mine may have a top size of 5 cm or larger. The top size of the coal to be employed in a fuel slurry is typically  $250\mu m$  or smaller. The particle size must be reduced, therefore, by a factor of at least 200; the final particle-size distribution must meet stringent requirements as noted in prior sections; and the comminution must be accomplished in an economical manner.

The process employed in the laboratory by the Atlantic Research Corporation starts with a hammer crusher with a cylindrical grating placed beneath the rotor, followed by a ball mill operated in the batch mode. The crushed product is introduced into the ball mill in either dry or slurry form.

The ball mill is one of only a number of different comminution devices which can be used to prepare pulverized coal. It has been selected for a number of reasons. First, it can achieve the desired results economically. Second, it is available in a range of sizes from laboratory mills to production mills with capacities of hundreds of tons per hour. Third, there exits a wealth of experimental data which enable one to predict performance, production costs, wear and machine life, and maintenance. Finally, grinding theories have been developed and compared with experimental ball mill results. These theories can be a useful aid in obtaining specific particle size distributions and will be discussed next.

#### Grinding Theory

A large body of literature exists on the theory of grinding, much of it emanating from Pennsylvania State University (5-7) and from the University of Utah (8). Most theories agree on a basic equation for batch grinding which can be written in a number of different ways. The form used in this work is most useful in obtaining results with a digital computer. It is convenient to treat the particle size distribution as a series of discrete sizes as would be obtained if ascertained by a series of screens. Consider a set of m screens, the coarsest screen being designated by 1 and the finest by m. The size of the i<sup>th</sup> screen is designated by its aperture  $\mathbf{x}_1$ . A constant screen ratio, q, is used so that  $\mathbf{x}_{1-1}/\mathbf{x}_1=\mathbf{q}$ . Material which is retained on screen i after passing through screen i-1 has a size range from  $\mathbf{x}_1$  to  $\mathbf{x}_{1-1}$  and has a weight  $W_1(t)$  at time t. This material will be referred to as being of size  $\mathbf{x}_1$ . The breakage equations representing weights in fractional form are:

$$\frac{dW_{i}(t)}{dt} = \sum_{j=1}^{i=1} b_{ij} k_{j} W_{j}(t) - k_{i} W_{i}(t)$$
(15)

$$W_{R}(t) + \sum_{i=1}^{m} W_{i}(t) = 1$$
 (16)

where  $W_R(t)$  = fractional weight passing finest screen  $k_i$  = rate constant =  $d \ln W_i / dt$ 

b. = breakage function = fraction of material obtained
 by primary breakage of size j retained on screen i.

In the general case,  $k_i$  is a function of both time and size  $x_i$ . The  $m^2$  values of  $b_{ij}$  are functions of the two sizes,  $x_i$  and  $x_j$ . In certain cases (which must be ascertained by comparison with experiment),  $k_i$  is independent of time and proportional to  $x_i$ , and  $b_{ij}$  is a normalized function which can be fit by the following equation:

$$b_{ij} = \left(\frac{x_{i-1}}{x_{j}}\right)^{y} - \left(\frac{x_{i}}{x_{j}}\right)^{y} , \qquad (17)$$

where y is an empirically determined constant

Combining the restricted equations for  $k_i$  and  $b_{i,j}$  with the differential equation, permits one to solve for particle size and particle size distribution as a function of time. Converting to finite difference form, and employing a fourth-order Runge-Kutta numerical solver, the equations have been programmed for a digital computer. Input values are the number of sieves, the screen ratio, q, the fractional weights initially on each screen, and the constant y. The output consists of the fractional weights on each screen at generalized times, a.t., where a is equal to the ratio  $k_i x_1/x_i$ .

Particle-size distributions obtained from ball mills often fit a Rosin- Rammler-Bennett (RRB) equation:

$$F_{p} = 1 - \exp \left[ -0.693 \left( \frac{D}{D_{50}} \right)^{\lambda} \right]$$
 (18)

where D = diameter  

$$\lambda$$
, D = constants  
 $D_{50}$  = 50% by volume of particles are smaller than this diameter

This distribution of particle diameters extends from zero to infinity, and, for practical purposes, must be truncated. In obtaining RRB fits of either computer results or experimental data, only points falling within the range  $0.05 < F_p < .97$  are used.

Typical results of particle-size distributions as predicted by the theoretical model are given in Figure 2. An empirical correlation of  $D_{50}$  values is shown in Figure 3. Predicted reciprocal  $D_{50}$  values are linear with time, based on the breakage function selected.

## Experimental Results

Coal-water slurries can be made from a wide range of coals. Although coals of lower ranks can be employed, bituminous coals have received the greatest attention. Typical coals have volatile contents between 25 and 40% and Hardgrove indices between 40 and 80. Approximately 30 such coals have been investigated thus far. Only a single coal will be discussed here; one with 35% volatile matter, 6% ash, and a Hardgrove index of 41. It can be considered a typical coal except for the grindability index. Considerable savings are obtained by using coals with larger Hardgrove indices.

Particle-size distribution curves are determined by wet sieving and by Fraunhofer plane diffraction for sub-sieve sizes. A Malvern Model 2200 particle sizer and Model 8T1800 analyzer is used for particles in the range of 1.2 - 118  $\mu$  m.

Prior to ball milling, the coal is crushed in a Holmes Bros. Model 201 crusher with grates containing 1.6 mm diameter holes. A size distribution curve for the product is shown in Figure 4 and 5. The curve fits the RRB equation with a D50 of 204  $\mu m$  and a value of 0.96 for  $\lambda$  . Values of  $\lambda$  are more meaningful when expressed as a ratio of D80/D50, the conversion equation being:

$$\frac{D_{80}}{D_{50}} = e^{.8424/\lambda} \tag{19}$$

For the crushed coal, the ratio  $D_{80}/D_{50}$  is equal to 2.4 and the  $D_{80}$  is  $491\mu m$ .

A jar mill without lifters, 21.3 cm in diameter, and 16.5 cm long is rotated at 60 rpm (70% of critical speed) with grinding media consisting of steel balls 1.59 cm in diameter. The empty volume of the jar is  $5800~\rm cm^3$ , the ball charge has a bulk volume of 2900 cm³ (50% loading), and the void volume within the ball bed is 1150 cm³. The volume of dry coal or slurry introduced as a charge to the mill is equal to the void volume, 1150 cm³.

The change in particle-size distribution with dry milling is shown in Figure 4. Figure 5 shows the results for wet milling in a slurry consisting of equal parts of coal and water and containing two parts of a petroleum-based surfactant per thousand parts of coal. These results were statistically fitted to the RRB equation; the resulting RRB curves are also shown in Figures 4 and 5. Using the RRB equations which best represent the data, the D50 values were calculated; reciprocal values are plotted against time in Figure 6. The D50 values as well as the distributions are well represented by the

theoretical milling equations with y = 2 for both wet and dry milling and with a = 4.0 s<sup>-1</sup> for dry milling and a = 9.3 s<sup>-1</sup> for wet milling. A comparison of the rate constants for wet and dry milling show that the former is the larger by a factor of more than two.

Although the milling results shown here are well-represented by simple breakage functions, such is not always the case. More complicated functions are required for different coals and different ball mill conditions.

## Obtaining The Optimum Distribution

One method of processing coal-water slurries is to blend dry milled coal with a slurry of wet milled coal. For example, combining two parts of dry milled coal with three parts of a 50% slurry results in a slurry containing 70% coal. A blend carried out in this ratio using the 50% slurry milled for 60 minutes and dry coal milled for 20 minutes has the particle-size distribution shown in Figure 7. A comparison is made with an optimum distribution calculated according to Equation 13 with Dp = 140, Ds = 2.5, and n = 0.24. The blend of the coarse dry milled coal (Ds0 = 56  $\mu$  m) and the finer wet-milled coal (Ds0 = 11  $\mu$  m) results in a near-optimum blend with a Ds0 = 19  $\mu$ m. This blend of a coarse and a fine grind to obtain an optimum distribution is sometimes called a bimodal blend even though that designation is more appropriately limited to distributions with two maxima.

## V. CONCLUSIONS

The optimum particle-size distributions for minimum viscosity or maximum packing density as derived by Furnas and by Farris have been shown to be equivalent in form, namely Equation 7. Furnas' equation for the exponent, n, differs somewhat from Farris' equation, the latter being preferred. These equations have been found to be very valuable in the formulation of coal-water slurries. It is not possible to predict the viscosity of these slurries from knowledge only of particle sizes because of the importance of surface chemistry and the influence on the chemistry of certain additives which which have been found useful in slurry development.

Near-optimum distributions of particle size can be obtained by blending products obtained by ball mills even though the individual distributions obtained from these mills are not optimum. Comminution in ball mills can be described theoretically; these techniques, derived from the literature, are useful in interpolating and extrapolating results.

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## VII. NOMENCLATURE

#### Particle-Size Distribution

- D = particle diameter
- f = crowding factor defined by Farris
- g = volume fraction of solids in a slurry
- $F_{\mathbf{p}}(\mathbf{D})$  cumulative volume of particles smaller than  $\mathbf{D}$
- N = number of component particle sizes
- R = ratio of absolute volume of particles present in consecutive size fractions.
- = diameter ratio between consecutive size fractions
- V = volume
- v = void volume
- $\phi$  = volume ratio defined by Farris, Equation 10

#### Subscripts

- i = designates size fractions; l is the smallest fraction
- designates largest diameter present
- s = designates smallest diameter present
- 50,80 = designate the volume percentage of particles smaller than specified diameter.

#### Grinding

- $b_{ij}$  = breakage function
- k = breakage rate constant
- m = number of sieves
- t = time
- $W_i$  = weight of material retained on the i<sup>th</sup> sieve
- $x_i$  = aperture of the i<sup>th</sup> sieve size
- y = constant in Equation 17
- $\lambda$  = constant in RRB distribution function, Equation 18

# Subscripts

i = designates sieves of different sizes, 1 being the largest sieve

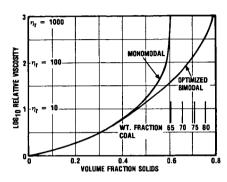


Figure 1. Effect of Bimodal Distribution on the Relative Viscosity of Suspensions of Mon-Interacting Spheres.

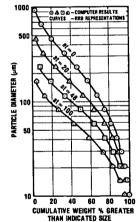


Figure 2. Particle Size Reduction in a Ball-Mill as Predicted by Theoretical Model y=2.

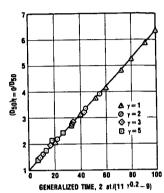


Figure 3. Empirical Correlation of  $\rm D_{50}$  Values of Ball-Milled Coal as Predicted by Theoretical Model.

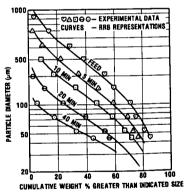


Figure 4. Experimental Particle Size Distributions Obtained in Dry Milling.

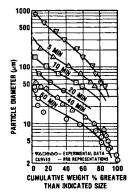
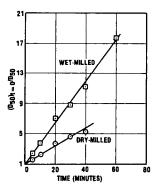


Figure 5. Experimental Particle Size Distributions
Obtained in Wet Milling.



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Figure 6. Correlation of Experimental D<sub>50</sub> Values — Batch Grinding in Laboratory Ball Mill.

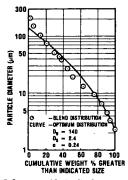


Figure 7. Comparison of Particle-Size Distribution of Blended Coal with Optimum Distribution.